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Are electron tweezers possible?

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ABSTRACT

Positively answering the question in the title, we demonstrate in this work single electron beam trapping and steering of 20–300 nm solid Al nanoparticles generated inside opaque submicron-sized molten Al–Si eutectic alloy spheres. Imaging of solid nanoparticles and liquid alloy in real time was performed using energy filtering in an analytical transmission electron microscope (TEM). Energy-filtering TEM combined with valence electron energy-loss spectroscopy enabled us to investigate *in situ* nanoscale transformations of the internal structure, temperature dependence of plasmon losses, and local electronic and optical properties under melting and crystallization of individual binary alloy particles. For particles below 20 nm in size, enhanced vibrations of the dynamic solid–liquid interface due to instabilities near the critical threshold were observed just before melting. The obtained results indicate that focused electron beams can act as a tool for manipulation of metal nanoparticles by transferring linear and angular mechanical momenta. Such thermally assisted electron tweezers can be utilized for touchless manipulation and processing of individual nano-objects and potentially for fabrication of assembled nanodevices with atomic level sensitivity and lateral resolution provided by modern electron optical systems. This is by three orders of magnitude better than for light microscopy utilized in conventional optical tweezers. New research directions and potential applications of trapping and tracking of nano-objects by focused electron beams are outlined.

1. Introduction

Understanding physical principles for controlling the dynamic behavior of nanoparticles in inhomogeneous gas (liquid)–solid systems is becoming increasingly important for nano-scale science and technology. Nondestructive trapping and manipulation of small particles in a liquid using a laser beam, which is refracted by the particle and transfers momentum to it, is known as a single-beam gradient force optical trap, or optical tweezers [1–3]. Optical tweezers have become a highly developed and heavily used tool of choice for many applications in biology, physics and chemistry, when gentle and remotely controllable manipulation of micro- and nano-objects is required. Nowadays, electromagnetic forces in optical tweezers are often employed to trap dielectric and metal particles ranging in size from tens of nanometers to several micrometers, and to manipulate them in all spatial directions [4–6]. Furthermore, 3D-trapping and orientation of individual Au nanorods using plasmon resonance has been performed [7] and holographic tweezers have been developed to trap tailored arrays of multiple micron-size objects [8,9]. The implementation of optical tweezers at a surface offers exciting opportunities for the elaboration of future lab-on-a-chip devices entirely operated with light. The transition from conventional 3D tweezers to 2D is made possible by exploiting evanescent fields bound at interfaces. In particular, stable trapping of single dielectric beads using surface plasmons (SP) under non-focused illumination has been recently demonstrated [10], whereas fine tuning of nanoparticle positions has been theoretically evaluated to be realizable by coupling to plasmonic nanostructures [11].

Our previous experiments utilizing partially molten submicron-sized Al–Si alloy spheres and a focused electron beam in a medium-voltage transmission electron microscope (TEM) [12–15] and numerical calculations of linear and angular momentum transfer from an electron beam to small particles in a scanning TEM (STEM) by others [16,17] indicate that optical trapping of particles by focused electron beams may occur. Although an experimental set-up is challenging, since it requires the trapping of particles in vacuum, TEM appears indeed as a promising technique to study optically trapped particles, providing an excellent spatial resolution (currently down to 48 pm) [18] when sub-nanometer electron beams are employed. Analytical electron microscopy (AEM) expands further potentialities of TEM for studying structural transformations of nanoscale materials by adding analytical capabilities such as energy-filtering imaging (EFTEM), electron energy-loss spectroscopy (EELS) and energy-

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dispersive X-ray spectrometry (EDXS), which allow one to probe local electronic and optical response properties, and more recently other physical (mechanical, transport) properties of nanostructured materials (see Ref. [19]). *In situ* AEM allows one to combine (S)TEM imaging and spectroscopy at up to sub-eV energy resolution with physical and chemical processing (e.g., heating/cooling, radiation damage experiments, and gas (vapor)–solid chemical reactions in environmental cells). These capabilities transform such instruments into a versatile micro- and nano-lab for high-spatial resolution analyses of various dynamic processes and fabrication of new materials and devices [14,15,20,21]. In this work, we demonstrate single electron-beam trapping and steering of solid nanoparticles inside individual opaque submicron-sized molten Al–Si eutectic alloy spheres during melting and crystallization of Al–Si eutectic alloy particles by EFTEM and valence EELS (VEELS). For this purpose, we employ the focused electron beam as a multifunctional probe for providing the following means in the course of experiments:

(a) fine temperature tuning during generation of solid Al nanoparticles inside submicron-sized molten Al–Si eutectic alloy spheres;
(b) monitoring nanoscale transformations of the internal structure, local electronic and optical properties under melting and crystallization of individual binary eutectic alloy particles, including plasmon imaging of solid metal particles inside opaque molten Al–Si alloy particles in real time;
(c) trapping, steering and re-melting of the Al nanoparticles.

2. Materials and methods

Atomized powder of Al-11.6 at% Si alloy, which has a eutectic (initial melting) temperature of 577° C and a liquidus (final melting) temperature of 581° C [22], was placed in a test tube of ethanol and dispersed using an ultrasonic cleaner. The test tube was removed from the cleaner and the larger particles were allowed to settle in the test tube for at least 1 h. The smaller particles (20–400 nm in diameter) present in the topmost liquid were collected in an eyedropper and deposited onto ultrathin carbon films supported by Cu-mesh grids. The 20–400 nm diameter particles of Al-11.6 at% Si alloy were examined in a JEOL[1] 2010 F Schottky field-emission analytical electron microscope. The instrument was equipped with a Gatan Model 678 Imaging Filter operated at 197 kV. EEL spectra were recorded at the energy resolution of 1.0 eV, based on the measured full-width at half-maximum of the zero-loss peak. Video recording of EFTEM images was carried out using a VCR with a time resolution of 1/30 s. Each frame was then imported into a Macintosh G4 computer with a video capture board and Adobe Premier 5.0 software. The particles were heated in the microscope using a Gatan double-tilt ($\pm 30^\circ$) heating holder with a maximum obtainable temperature of 800 °C. Real-time plasmon imaging of melting/crystallization of Al nanospheres inside the submicron-sized Al–Si alloy particles was performed with a 6–10 eV energy window centered at the first Al plasmon peak at 15 eV.

3. Results and discussions

In the following sections, we first discuss melting of the Al-11.6 at% Si alloy particles and observation of the changes that occur inside the particles during this process using EFTEM. We then examine the effect of the melting transition on the EEL spectra and optical properties of the nanoparticles. Understanding these behaviors is important for interpretation of the ability to use the electron beam as an "electron tweezer" to steer a solid Al nanoparticle inside molten Al–Si alloy liquid. This process is then demonstrated experimentally and followed by an analytical analysis, which explains the gradient force necessary for such a process to occur. Lastly, further potential possibilities of using such electron tweezers are described.

3.1. EFTEM/VEELS of melting of Al–Si alloy particles

In this part, we discuss an investigation of the melting of the Al-11.6 at% Si eutectic alloy submicron and nano-sized particles *in situ*. When the temperature was increased just below the liquidus of 581° C [22], the particles partially melted to form a stable two-phase solid–liquid mixture, consisting mainly of a spherical-shaped solid α-Al nanoparticle inside a spherical shell of liquid Al–Si alloy, all of which is contained inside the submicron-sized particles by a thin aluminum-oxide shell on the particle surface [12,23]. Zero-loss filtering often improves the contrast and resolution of the particle structures as compared to conventional TEM imaging because blurring and chromatic aberration due to inelastically scattered electrons are eliminated. However, internal structural details for the particle shown in the zero-loss (elastic electron) image (Fig. 1, 0 ± 5 eV) are hardly visible due to its thickness of 223 nm. Contrast tuning using energy filtering at selected energy losses provided better phase separation of the Al-rich areas from 10 to 20 nm-sized Si-rich precipitates using the Al volume plasmon (VP) losses (15 ± 5 eV) and allowed identification of the non-uniform oxide shell 5–15 nm thick at oxide plasmon losses (25 ± 5 eV) (Fig. 1). The oxide shell contains the Al-rich liquid in the particle when it is partially or fully molten and undergoes creep to relieve the high stress on the order of 15 GPa

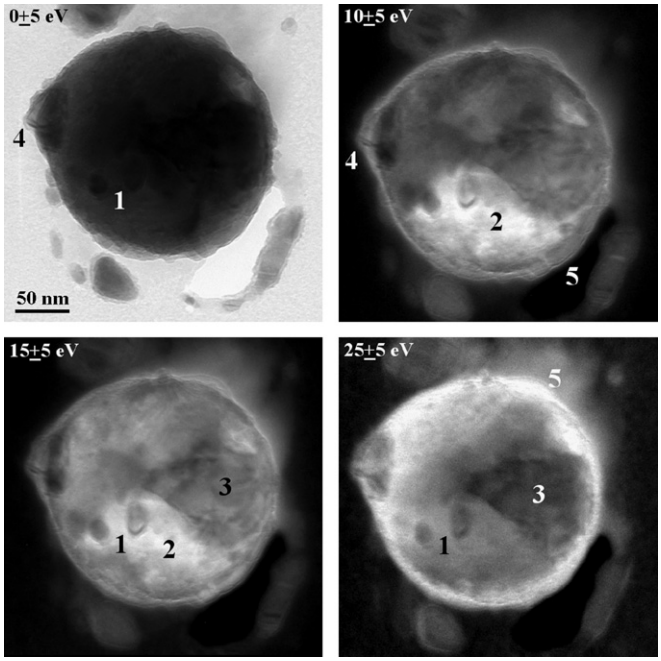

Fig. 1. A molten Al-11.6 at% Si particle at $T=656$ °C. EFTEM, a 10 eV window centered at 0, 10, 15 and 25 eV energy losses. Contrast tuning under energy filtering in the range between 0 and 25 eV reveals structural details of a molten single-crystal α-Al matrix with multiple embedded 10–20 nm Si precipitates (1), α-Al-rich area (2), Si-rich area (3), defect cavern (4), as a result of ejection of liquid through a 5–15 nm-thick oxide shell containing the alloy (5), and its rupture during melting.

[1] Certain commercial equipment, instruments, or materials are identified in this document. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products identified are necessarily the best available for the purpose.